Growth of High Quality Three Dimensional Topological Insulator Bi2-xSbxTe3-ySey Thin Film by Physical Vapor Deposition and Fabrication of Topological p - n Junction

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論　文　内　容　要　旨

氏　　名	TU NGOC HAN	提出年	平成 28 年
学位論文の題　　目	**Growth of High Quality Three Dimensional Topological Insulator $Bi_{2-x}Sb_xTe_{3-y}Se_y$ Thin Film by Physical Vapor Deposition and Fabrication of Topological p - n Junction.**		

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INTRODUCTION

Three –dimensional topological insulators (3D – TIs) have been attracted a lot of attentions owing to their time – reversal – symmetry – protected surface states. They are of interest for both fundamental and application researches because of special properties related to the electrically insulating bulk states and the spin – helical surface states (1-3). Although a huge of interests and research activities in 3D – TIs were envisioned in recent years, there were many experimental problems in the real materials to realize the theoretical expectations, due to the lack of insulating bulk states. One of the most promising solutions to solve this problem would be implementation of TI nanostructures with the bulk insulating states which is expected to enhance the contribution of the surface states in the electrical transport. Once the surface states are dominant, the novel electronic transport could be observed.

In this thesis, I report growth methods of high quality $Bi_{2-x}Sb_xTe_{3-y}Se_y$ (BSTS) ultrathin film by the vapor-liquid-solid (VLS) growth which is known as one of the best material for 3D - TI research. Using present high quality ultrathin film, electrical transport properties of BSTS ultrathin film as well as the Klein tunneling, one of the most interesting feature of topological surface states, were experimentally investigated.

EXPERIMENTS

A growth method of BSTS ultrathin film was developed using a modified vapor – solid deposition. The main modification was to employ an inner tube in the growth system (Fig.1a). This setting allowed us to grow very large – size and controllable thickness film on mica substrates. Characterization results of BSTS ultrathin films, including SEM-EDX, AFM, X-ray diffraction, Raman spectroscopy, revealed that high – quality ultrathin films with the appropriate chemical composition and the crystal structure were grown. Importantly, a thickness of the BSTS ultrathin film could be precisely controlled in the range of 3 – 50 nm with 1 cm^2 full covered on mica substrate by a low - cost and facile preparation method (Fig. 1b). Moreover, I developed the transfer method, which can peel off a BSTS film from mica substrate and then transfer on any arbitrary substrates. Using this technique, BSTS film grown on mica substrate in the centimeter scale can be transferred to SiO_2/ Si substrate with any thickness, down to 2 ~ 3 nm while keeping the morphology as well as composition (Fig.1c, d). Raman spectroscopy of BSTS films with various thicknesses showed the same peak positions from a bulk single crystal to 3 nm film, indicating that present thin films were composed of a same composition.

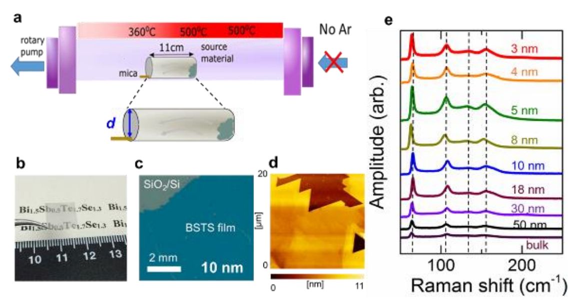

Figure 1 (a) Schematic view illustrating the growth set up system. (b) Photograph image of 10 nm BSTS film with full coverage on mica. (c) Optical images of (a) after transfer onto SiO_2/ Si substrate. (d) AFM image of the 10 nm film transfer on SiO_2/ Si substrate. (e) Raman spectroscopy of BSTS films with various thicknesses.

GROWTH OF LARGE – SIZED BSTS ULTRATHIN FILM

Morphology of BSTS thin film was found to strongly depend on the inner tube diameter (d). By using d = 1.4 cm, a film thickness was precisely controlled by growth time from 3 nm to 50 nm, with a step of 1 nm. Fig. 2 showed the scanning electron microscopy (SEM) images of BSTS films. A BSTS film composed of well oriented BSTS NPs and gradually increased by prolonging the time growth. Therefore, controlling the growth time, we are able to adjust the film thickness.

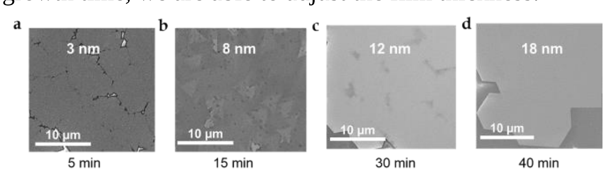

Figure 2. (a – d) Scanning electron microscopy (SEM) images of BSTS films for the inner tube diameter d = 1.4 cm against the growth time.

ELECTRICAL TRANSPORT OF ULTRATHIN BSTS FILMS

Temperature and thickness dependences of electrical transports for as – grown BSTS films with various thicknesses were shown in Fig. 3 (a). By changing a film thickness, a surface contribution was expected to emphasize, resulting in the semiconductor – metallic transition of R_{sheet} (T) in the range of 50 – 8 nm. From 5 – 3 nm, R_{sheet} (T) showed the insulating behavior, which maybe come from a hybridization gap due to the tunneling between top and bottom surfaces. The existence of π Berry phase was confirmed by analyzing Shubnikov de Haas (SdH) oscillations of an 18 nm film, as shown in Fig. 3 (b, c, d).

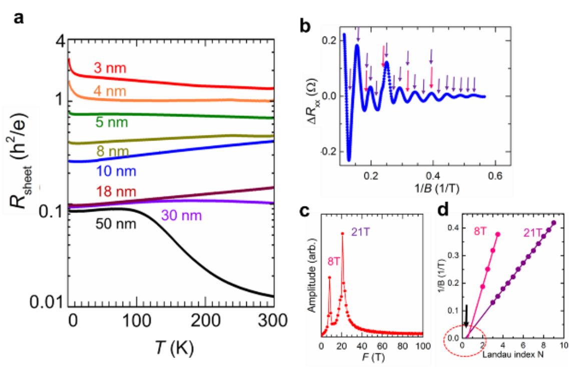

Figure 3. Electrical transport properties of BSTS films. (a) Temperature dependence of R_{sheet} (T) with various thicknesses (b) SdH oscillations observed in a 18 nm thickness film at 2 K. The arrows indicated the peak and valley positions of oscillation with 8 T and 21 T frequencies, based on (c) FFT results. (d) Landau Level fan diagram constructed form the analysis of the ΔR_{xx} data, here, integer index N (half integer N +1/2) are assigned to the minima (maxima). Making a linear fitting to data, the straight – line fitting extrapolates to 0.5 ± 0.03 for oscillation with 8T frequency and 0.41 ± 0.05 for oscillation with 21 T frequency, which are close to the value ½ expected for Dirac fermions.

IN – PLANE TOPOLOGICAL P – N JUNCTION IN 3D – TI BSTS

Considering the novel electric transport for the topological p-n junction (TPNJ) as well as its applications to 3D-TI devices by employing ultralow dissipative spin and charge currents, a truly firm confirmation and understanding on TPNJ fabricated on the in-plane surface of 3D-TIs are desired (4-5). A 10 nm thickness BSTS film was firstly transferred on SiO_2/Si (300 nm), then F4-TCNQ, electron accepter molecules, were deposited on a half of film (Fig. 4 (b)). Due to a charge transfer process between F4 – TCNQ and BSTS, a region covered by F4-TCNQ was changed to p – type on the top surface, forming a p – n junction on the top surface of sample. The Fermi level of the bottom surface was controlled by bottom gate, and the topological p – n junction for both top and bottom surface was formed. Fig.4 (c) showed an evolution of four probe resistance (R_{xx}) across the TPNJ made on the top surface between pristine BSTS (n-type surface) and F4-TCNQ/BSTS (p-type surface) as a function of V_G. In the range of V_G between -90 and -30 V, where TPNJ states could be created both on the top and the bottom surfaces, R_{xx} showed a markedly abrupt and step-like jump at two particular gate voltages: from 12 to 18 kΩ at V_G = -30 V and from 14 to 28 kΩ at V_G = -90 V. These values were extremely high compared with the values of R_{xx} where no TPNJ at the interface between BSTS and F4-TCNQ/BSTS was created. A clear enhancement in R_{xx} observed in the TPNJ in the FET structure provides important and direct experimental evidence that the spin-locked TPNJ strongly scatters electron transmission, which is consistent with theoretical expectation.

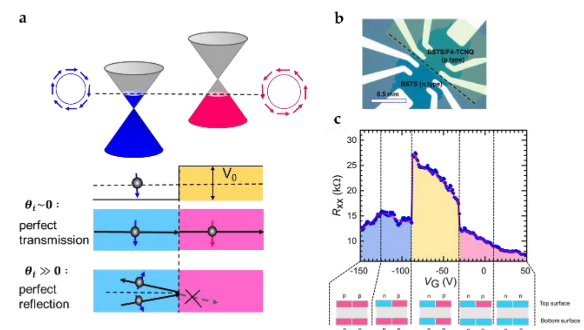

Figure 4. Electric transport of TPNJ device at 2 K. (a) A schematic for topological p – n junction. (b) Microscope view of a topological p-n junction device. (c) A gate voltage (V_G) dependence of a four probe resistance (R_{xx}) across the TPNJ made between pristine BSTS and BSTS/F4-TCNQ.

CONCLUSION

The growth of BSTS on a mica substrate followed by the nondestructive film transfer process can accurately manipulate centimeter-size ultrathin films of high quality 3D-TI BSTS. Employing these high quality BSTS ultrathin films, the electrical transport properties of BSTS ultrathin film as well as the first observation of the intrinsic electrical transport of TPNJ were obtained. Results of TPNJ can provide a deep insight into the new type of planar TPNJ, which may also be a base for further studies.

References

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別　紙

論文審査の結果の要旨

3次元トポロジカル絶縁体は、バルクの電子状態が非自明なトポロジカル不変量により規定される物質群である。この非自明なバルクの電子状態が真空に接続する際、界面にヘリカルにスピン偏極したディラックコーンが出現することが知られており、その特異な電荷とスピンの輸送現象に基づく新奇な物性の舞台として注目されている。本論文は、n型の 3 次元トポロジカル絶縁体$Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$(BSTS)を対象として、高品質・大型薄膜の育成とトポロジカル表面状態に特徴的なスピンと電荷の輸送現象の観測に関するものである。

トポロジカル表面状態の輸送現象の観測においては、高いバルク絶縁性と薄膜化が重要である。BSTS は、高いバルク絶縁性を示し、さらに表面状態のディラック中性点がバルクバンドギャップ内に位置することから、表面状態に特徴的な輸送現象の観測に最適な物質である。一方で、この4元合金の組成と膜厚を同時に精密に制御した薄膜の育成はこれまでなされていなかった。本研究では、物理気相輸送法を用いることで、組成と膜厚が制御可能な大型・高品質 BSTS 薄膜の育成手法を確立すると共に、様々な基板への薄膜の転写を可能とした。電子輸送現象の実験により、輸送現象に関係する膜厚に依存した表面状態の制御、超薄膜領域での表面と裏面の波動関数混成によるトポロジカル表面状態におけるエネルギーギャップの形成、トポロジカル表面状態に特徴的な両極性の電子状態とベリー位相による低散逸な伝導状態を観測し、形成した BSTS 薄膜が高い電子易動度を保った状態であることを示した。さらに、有機半導体 F4-TCNQ 分子の局所的な界面形成による電荷移動と電界効果トランジスタ構造を用いた電界効果を併せることでトポロジカル p-n 接合を作製し、トポロジカル表面状態のカイラルトンネル状態として理解できる直流抵抗の急激な増大を初めて示した。

本論文は、3 次元トポロジカル絶縁体の新奇な物性の発現とデバイス応用において重要な化合物である BSTS において、(1)高品質・大型薄膜の育成(2)デバイス作製技術の確立(3)薄膜の基礎物性の解明並びにトポロジカル表面状態に特徴的なスピンと電荷の輸送現象の観測を行ったものである。この成果は、申請者が高度の学識と自立して研究する能力があることを示すと判定される。よって、Tu Ngoc Han (ダウ ノック ハン)氏提出の博士論文は、博士（理学）の学位論文として合格と認める。